



PCT/GB 2003/001196

REC'D 11 APR 2003

WIPO

PCT

INVESTOR IN PEOPLE

PRIORITY DOCUMENT
SUBMITTED OR TRANSMITTED IN
COMPLIANCE WITH
RULE 17.1(a) OR (b)

The Patent Office
Concept House
Cardiff Road
Newport
South Wales
NP10 8QQ

I, the undersigned, being an officer duly authorised in accordance with Section 74(1) and (4) of the Deregulation & Contracting Out Act 1994, to sign and issue certificates on behalf of the Comptroller-General, hereby certify that annexed hereto is a true copy of the documents as originally filed in connection with the patent application identified therein.

In accordance with the Patents (Companies Re-registration) Rules 1982, if a company named in this certificate and any accompanying documents has re-registered under the Companies Act 1980 with the same name as that with which it was registered immediately before re-registration save for the substitution as, or inclusion as, the last part of the name of the words "public limited company" or their equivalents in Welsh, references to the name of the company in this certificate and any accompanying documents shall be treated as references to the name with which it is so re-registered.

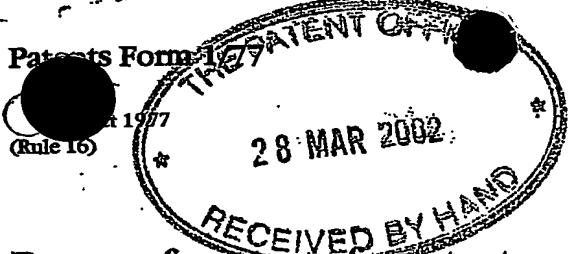
In accordance with the rules, the words "public limited company" may be replaced by p.l.c., plc, P.L.C. or PLC.

Re-registration under the Companies Act does not constitute a new legal entity but merely subjects the company to certain additional company law rules.

Signed

Dated 11 March 2003

Stephen Honolley



30MAR02 E707560-1 002093
P01/7700 0.00-0207438.3

Request for grant of a patent

(See the notes on the back of this form. You can also get an explanatory leaflet from the Patent Office to help you fill in this form)

The Patent Office

Cardiff Road
Newport
South Wales
NP10 8QQ

1. Your reference

PPD 50679/GB/P

2. Patent application number

(The Patent Office will fill in this part)

0207438.3

28 MAR 2002

3. Full name, address and postcode of the or of each applicant (underline all surnames)

8330 748001

SYNGENTA Limited
European Regional Centre
Priestley Road
Surrey Research Park, Guildford,
Surrey, GU2 7YH, United Kingdom

Patents ADP number (if you know it)

—6254007002—

If the applicant is a corporate body, give the country/state of its incorporation

UNITED KINGDOM

4. Title of the invention

FORMULATION

5. Name of your agent (if you have one)

"Address for service" in the United Kingdom to which all correspondence should be sent (including the postcode)

Michael James RICKS
Intellectual Property Department
Syngenta Limited
Jealott's Hill International Research Centre
PO Box 3538
Bracknell, Berkshire, RG42 6YA
UNITED KINGDOM

Patents ADP number (if you know it)

814 829 8002

6. If you are declaring priority from one or more earlier patent applications, give the country and the date of filing of the or of each of these earlier applications and (if you know it) the or each application number

Country Priority application number
(if you know it) Date of filing
(day / month / year)

7. If this application is divided or otherwise derived from an earlier UK application, give the number and the filing date of the earlier application

Number of earlier application Date of filing
(day / month / year)

8. Is a statement of inventorship and of right to grant of a patent required in support of this request? (Answer 'Yes' if:
a) any applicant named in part 3 is not an inventor, or
b) there is an inventor who is not named as an applicant, or
c) any named applicant is a corporate body.
See note (d))

YES (b)

9. Enter the number of sheets for any of the following items you are filing with this form.
Do not count copies of the same document

| | |
|----------------------------------|-------|
| Continuation sheets of this form | 09 |
| Description | 01 DM |
| Claim(s) | 01 |
| Abstract | 01 |
| Drawing(s) | 00 |

10. If you are also filing any of the following, state how many against each item.

Priority documents

Translations of priority documents

Statement of inventorship and right to grant of a patent (Patents Form 7/77)

Request for preliminary examination and search (Patents Form 9/77)

Request for substantive examination (Patents Form 10/77)

Any other documents
(Please specify)

11.

I/We request the grant of a patent on the basis of this application.
Syngenta Limited

Signature
Authorised Signatory

Date

28/3/02

12. Name and daytime telephone number of person to contact in the United Kingdom

Joanna Carmen CHANDLER = 01344 414365

Warning

After an application for a patent has been filed, the Comptroller of the Patent Office will consider whether publication or communication of the invention should be prohibited or restricted under Section 22 of the Patents Act 1977. You will be informed if it is necessary to prohibit or restrict your invention in this way. Furthermore, if you live in the United Kingdom, Section 23 of the Patents Act 1977 stops you from applying for a patent abroad without first getting written permission from the Patent Office unless an application has been filed at least 6 weeks beforehand in the United Kingdom for a patent for the same invention and either no direction prohibiting publication or communication has been given, or any such direction has been revoked.

Notes

- If you need help to fill in this form or you have any questions, please contact the Patent Office on 08459 500505.
- Write your answers in capital letters using black ink or you may type them.
- If there is not enough space for all the relevant details on any part of this form, please continue on a separate sheet of paper and write "see continuation sheet" in the relevant part(s). Any continuation sheet should be attached to this form.
- If you have answered 'Yes' Patents Form 7/77 will need to be filed.
- Once you have filled in the form you must remember to sign and date it.
- For details of the fee and ways to pay please contact the Patent Office.

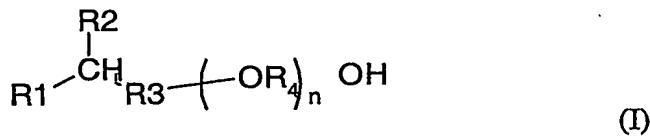
FORMULATION

This invention relates to a formulation and in particular to a formulation of the herbicide glyphosate.

5 Formulations of the herbicide glyphosate are generally available in the form of a concentrate which is diluted with water by the user prior to application. For large-scale agricultural use the dilution of the concentrate normally takes place in a spray tank from which the diluted product is applied through a suitable spraying apparatus. A common problem encountered is that of excessive foaming of the concentrate. Many glyphosate 10 formulations therefore contain commercially available anti-foam agents designed to minimise this problem. Anti-foam agents are normally effective at low concentration (typically of the order of 1 g/l). This concentration is much lower than that of bioperformance enhancing adjuvants which may also be present in the formulation (typically about 100 to 150 g/l), anti-foam agents are normally assumed to have little or no effect on 15 bioperformance.

We have now found that certain polyethyleneoxide alkanols are effective as anti-foam agents and, despite being present at relatively low concentration, additionally have surprising beneficial effects on bioperformance.

According to the present invention there is provided a herbicidal composition 20 comprising glyphosate and a poly(alkylene oxide) alkanol having the formula



wherein R_1 and R_2 are methyl or ethyl, R_3 is a straight chain alkylene group containing from 5 to 12 carbon atoms, R_4 is an alkylene group containing 2 or 3 carbon atoms and n is from 4 to 20.

25 The group $(\text{OR}_4)_n$ is a poly (ethylene oxide) group, poly (propylene oxide) group or a mixed poly (ethylene oxide/propylene oxide) group. In the case of compounds having a range of degrees of alkoxyoxylation, n may be an average number. It is preferred that n is from 6 to 14, for example from 6 to 10. An especially preferred value of n is about 8.

30 R_1 and R_2 are preferably methyl. R_3 preferably contains from 7 to 11 carbon atoms and an especially preferred group R_3 is $-(\text{CH}_2)_{10}-$.

A preferred poly(alkylene oxide) alkanol of formula (I) is ethoxylated, propoxylated or mixed alkoxylated/propoxylated iso-tridecyl alcohol wherein the degree of alkoxylation is from 6 to 14.

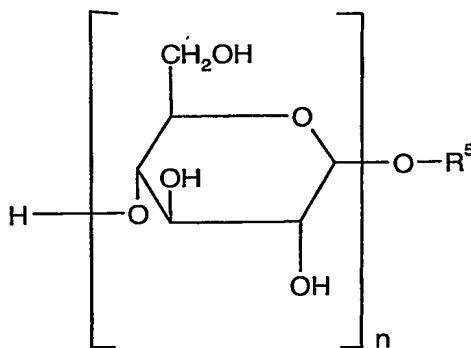
Glyphosate is normally used in the form of a water-soluble salt of glyphosate acid (N-phosphonomethylglycine) and the term glyphosate as used herein includes glyphosate acid and its salts, esters and derivatives. Typical glyphosate salts include isopropylamine, trimethylsulphonium, monoethanolamine, ammonium, potassium and sodium. As indicated previously, the present invention is particularly relevant to the formulation of glyphosate concentrates. Such concentrates typically contain of the order of 240 g/l based on glyphosate acid, although there is a move towards more concentrated formulations for example from 240 to 400 g/l and more particularly from 340 to 380 g/l based on glyphosate acid

It is a particular feature of the present invention that the compound of formula (I) operates both as an anti-foam and as a bioperformance enhancing agent at lower concentrations than are conventional for bioperformance enhancing adjuvants. Thus whilst higher proportions of the compound of formula (I) may be used, the benefits of the invention are most apparent when the concentration of the compound of formula (I) is below 100 g/l, for example from 0.1 to 100 g/l. A concentration of the compound of formula (I) of at least 1 g/l is preferred, for example from 1 to 50 g/l and especially from 1 to 20 g/l.

It is a further advantage of the present invention that the compound of formula (I) remains fully dispersed on storage of the composition. In contrast certain commonly used commercial anti-foams tend to separate out from the composition on storage and thereby lose their effectiveness, particularly if the concentrate is re-distributed from bulk storage into smaller containers.

It will be appreciated that relatively low levels of the compound of formula (I) are unlikely to provide sufficient bioperformance enhancement on their own and it is preferred that the compound of formula (I) is used with one or more additional bioperformance enhancing adjuvants. A wide range of bioperformance enhancing adjuvants suitable for use with glyphosate are well known in the art. We have found that the combination of a compound of formula (I) with an alkylpolyglycoside adjuvant provides an unexpectedly high level of bioperformance enhancement of glyphosate combined with an effective anti-foam action, particularly with respect to foaming of the concentrate. The effect is of such a magnitude that it is believed that there is a beneficial interaction or synergy between the compound of formula (I) and the alkylpolyglycoside.

The alkylglycoside for use in the present invention may be obtained by the reaction of alkanols with glucose or other mono- or di- or polysaccharides. As used herein the term "alkylglycoside" includes an alkylmonoglycoside and an alkylpolyglycoside. Preferred alkylglycosides for use in the present invention are alkylglucosides obtained by the reaction of glucose with a straight or branched chain alkanol or mixture of alkanols, for example a mixture of alkanols containing 7 to 18, preferably 7 to 16 carbon atoms for example 8 to 10 carbon atoms. The number of glucose groups per alkyl group in the molecule may vary and alkyl mono- or di- or polyglucose or saccharide derivatives are possible. Commercial alkylpolyglucosides usually contain a mixture of derivatives having an average number of glycoside groups per alkyl group. Thus alkylglycosides have the general formula (III)



(III)

wherein n is the degree of polymerisation and is typically within the range from 1 to 3, for example from 1 to 2, and R⁵ is a branched or straight chain alkyl group having from 4 to 18 carbon atoms or a mixture of alkyl groups having an average value within the given range. Typical of alkylglycosides is the product commercially available under the trade names AL2042 and AL2575 (Uniqema) and AGRIMUL PG2067 (Henkel Corp) wherein n is an average of 1.7 and R⁵ is a mixture of octyl (45%) and decyl (55%), the product commercially available under the trade name AGRIMUL PG2069 (Henkel Corp) wherein n is an average of 1.6 and R⁵ is a mixture of nonyl (20%), decyl (40%) and undecyl (40%) and the product commercially available under the trade name BEROL AG6202 (Akzo Nobel) which is 2-ethyl-1-hexylglycoside.

The additional biopreformance enhancing adjuvant, for example the alkylpolyglycoside, is suitably used at conventional concentrations of from about 80g/l to about 250g/l, for example from about 100g/l to about 150g/l and the ratio of the compound of formula (I) to the second biopreformance enhancing adjuvant is preferably from about 1 : 1 to 1 : 100 and more preferably from 1 : 10 to 1 : 40.

The compound of formula (I) may be used as the sole anti-foam agent or may be used in combination with or as a partial replacement for another anti-foam agent.

Other conventional additives such as humectants, activity enhancers (such as inorganic ammonium salts), anti-freeze agents, wetters, or other additional surfactants may 5 be added if desired. Similarly, additional water-soluble herbicides or other agrochemicals such as fungicides and insecticides may be incorporated if desired but the present invention is primarily concerned with compositions in which the only active agrochemical ingredient is glyphosate.

When diluted for use, compositions of the present invention are active against a broad 10 range of weed species including monocotyledonous and dicotyledonous species. The compositions are suitably applied directly to unwanted plants (post-emergence application).

Thus according to a further aspect of the present invention there is provided a process of severely damaging or killing unwanted plants which comprises diluting a concentrated 15 composition of the present invention and applying to the plants a herbicidally effective amount of a said diluted composition.

The rate of application of the composition of the invention will depend on a number of factors including, for example, the identity of the plants whose growth is to be inhibited and whether the compound is to be applied for foliage or root uptake. As a general guide, however, an application rate of from 0.001 to 20 kilograms per hectare is suitable while from 20 0.025 to 10 kilograms per hectare may be preferred.

The present invention is illustrated in its application to the formulation of ammonium glyphosate. Ammonium glyphosate is a particularly suitable salt for use in high-strength glyphosate concentrates in which an effective proportion of a surfactant system and inorganic adjuvant is "built-in" to the composition. The term "built-in" as used herein 25 indicates a composition in which the required primary adjuvants are contained within a physically stable concentrate composition and do not have to be added during the tank mix stage. This does not of course preclude the operator adding further adjuvants during tank mix if desired, but there is no necessity for the addition of further adjuvants. The difficulties of providing effective high-strength glyphosate concentrates containing adjuvant systems are 30 considerable, since such concentrates have to be physically stable over extended storage at the possible extremes of ambient temperatures likely to be encountered in commercial usage.

As used herein, the term "high-strength" aqueous glyphosate concentrate indicates a concentrate in which the glyphosate concentration is greater than 240 g/l based on

glyphosate acid content, for example from 240 to 400 g/l. An especially preferred concentration is from 340 to 380 g/l based on glyphosate acid. It should be noted that, unless otherwise stated, all concentrations of glyphosate are given herein in terms of the percentage by weight of glyphosate acid even when the glyphosate anion is balanced by an ammonium cation.

The ammonium glyphosate composition may alternatively be thought of as a combination of the ammonium salt of glyphosate, an ammonium salt such as ammonium sulphate or diammonium phosphate and the alkylglycoside. It will be appreciated however that once in solution, the composition is characterised by a total ammonium cation content balanced by glyphosate anion and, for example sulphate anion. It is convenient nevertheless to express the content of the formulation in terms of ammonium salt content and ammonium glyphosate content. The ammonium glyphosate composition of the present invention provides a physically stable composition even at a high loading of ammonium salt, for example ammonium sulphate, without having to reduce either the surfactant concentration or the glyphosate concentration. This is particularly surprising in view of the high electrolyte content of the composition. Thus to achieve a satisfactory enhancement of the activity of glyphosate, it is desirable to incorporate more than 70 g/l ammonium salt and preferably more than about 80 g/l. The upper limit of content of ammonium salt will depend on the desired content of surfactant and glyphosate ion and whilst levels as high as 180 g/l may be used if desired, the practical upper limit of the content of ammonium salt will normally be 160 g/l, or more preferably 150 g/l. Especially effective results are obtained with a content of ammonium salt of from 80 to 140 g/l since a surprisingly high and effective loading of glyphosate ion and alkylglycoside surfactant respectively may be obtained in such compositions.

As noted above, whilst the ammonium ion present in the aqueous ammonium glyphosate composition should not be thought of as being associated specifically with either with the glyphosate anion or for example the sulphate anion, it is convenient to express the concentration of ammonium ion relative to glyphosate ion as being in addition to that present in the "ammonium salt" such as ammonium sulphate. Thus the molar ratio of ammonium ion (in excess of that in the "ammonium salt") to glyphosate anion is preferably from 1.5 : 1 to 2 : 1, for example from 1.7 : 1 to 1.9 : 1. Particularly effective results are obtained at a ratio of (excess) ammonium ion to glyphosate ion of about 1.9 : 1.

As noted above, the content of the alkylglycoside surfactant system in the aqueous concentrate is generally from about 80 to 250 g/l. The upper limit of 250 g/l is more appropriate when relatively lower levels of ammonium salt are used whilst the lower limit of 80 g/l is more appropriate when relatively higher levels of ammonium salt are used.

5 Preferably therefore when the content of the ammonium salt content is from about 80 to about 100 g/l, the content of the alkylglycoside is from about 250 g/l to about 200 g/l, for example from about 245 g/l to about 210 g/l. When the content of the ammonium salt is from about 100 g/l to about 150 g/l, the content of the alkylglycoside is from about 140 g/l to about 200 g/l, for example about 140 g/l to about 175 g/l.

10 It is a particular advantage of the ammonium glyphosate composition of the present invention that a single surfactant, the alkylglycoside may be used as the sole additional bioperformance enhancing agent with the compound of formula (I), thus providing processing and supply advantages.

15 If desired however, a proportion of the alkylglycoside may be replaced by a an alkoxylated alkylamine and in some circumstances a further increase in biological activity may be observed thereby. The phrase "alkylglycoside surfactant system" as used above includes both the alkylglycoside and the alkoxylated alkylamine, if used and the preferred concentrations given above thus include both the alkylglycoside surfactant and the alkoxylated alkylamine, if used.

20 The ratio of alkylglycoside to alkoxylated alkylamine in the alkylglycoside surfactant system is preferably from about 1 part by weight of alkylglycoside per 1 part by weight of alkoxylated alkylamine to about 10 parts by weight of alkylglycoside per 1 part by weight of alkoxylated alkylamine. An especially preferred ratio is from about 5 to about 8 parts by weight of alkylglycoside per 1 part by weight of alkoxylated alkylamine.

25 Suitably the alkyl group in the alkoxylated alkylamine contains from 8 to 22 carbon atoms (or an average of from 8 to 22 carbon atoms if a mixture of alkyl groups is present) and may be linear or branched. It is especially preferred that the alkyl group contains from 10 to 20 carbon atoms. Specific examples of preferred alkoxylated alkylamines are alkoxylated derivatives of cocamine, tallowamine, oleylamine and stearylamine. Typically such alkoxylated alkylamine surfactants are available with an average degree of alkoxylation of from 1 to about 15. Suitable alkoxy groups include ethoxy, propoxy or a mixture thereof. Ethoxy is particularly preferred.

Whilst it is possible to use an alkoxylated alkylamine in the composition of the present invention having an average degree of alkoxylation (or more specifically ethoxylation) in the commercially available range, for example from 1 to about 15, we have found that ethoxylated alkylamines having a high degree of ethoxylation are less compatible with the high-strength composition than are those having a lower degree of ethoxylation. Thus it is preferred that the average degree of alkoxylation (or more specifically ethoxylation) is from 2 to 12. Thus it is especially preferred that the average degree of alkoxylation (or more specifically ethoxylation) is from 2 to 5. An example of a particularly suitable alkoxylated alkylamine is an ethoxylated alkylamine having an average degree of ethoxylation of about 2, for example an ethoxylated cocoamine having an average degree of ethoxylation of about 2.

Whilst advantages may be obtained through the replacement of a proportion of the alkylglycoside with an alkoxylated alkylamine, such advantages are offset by an increase in viscosity of the composition.

If the viscosity of the composition is high, for example if an alkoxylated alkylamine is used, it may be appropriate to add a viscosity modifying agent. Suitable viscosity modifying agents include propylene glycol.

The compositions of the present invention may be made by mixing the components in the desired proportions. The particular combination of ions in the composition of the present invention may be obtained from a range of different starting materials. The order of addition is not particularly important. Thus for example solid diammonium glyphosate and glyphosate acid (to give the desired ammonium to glyphosate ion ratio) may be added with solid ammonium sulphate into water into which the alkylglycoside has been dissolved. Alternatively ammonia may be added to glyphosate acid in the presence of ammonium sulphate or alternatively still, ammonia may be added to glyphosate acid in the presence of sulphuric acid.

Compositions of the present invention may also be provided in a diluted and ready-to-use form. Additional adjuvants suitable for ready-to-use formulations may also be added e.g. antifreeze, polymers and dyes.

The invention is illustrated by the following Examples in which all parts and percentages are by weight unless otherwise stated.

EXAMPLE 1

5 In the following Examples, compositions were prepared by adding ammonia to neutralise glyphosate acid (PMG acid) in the form of an aqueous slurry in order to generate the desired ratio of ammonium ion to glyphosate (1.9 : 1). The remaining components were added in the indicated proportions (% w/w) and water was added proportionately to make up the balance. The compound of formula (I) was GENAPOL X080, iso-tridecyl alcohol having 8 polyethylene oxide units (n in formula (I) is 8). The alkylpolyglycoside was AL2575, from UNIQEMA.

10 A comparison was prepared in which no compound of formula (I) was used. The quantity of alkylpolyglycoside was increased proportionately and additional anti-foam was added.

Both the composition of the invention and the comparison with added conventional antifoam had satisfactory and essentially equivalent foaming characteristics.

15 In the following Table, compositions are given as % w/w at a composition density of 1.28 g/cm³:

| | Example 1 | Comparison |
|---|-----------|------------|
| Glyphosate acid (as technical wet paste) | 28 | 28 |
| Ammonia (anhydrous) | 8.2 | 8.2 |
| Sulfuric acid (96% Commercial) | 8.5 | 8.5 |
| AL2575 | 13.0 | 13.4 |
| Antifoam MSA | 0.05 | 0.05 |
| Fluowet PL80 | 0 | 0.055 |
| Isotridecyl alcohol 8EO | 5.0 | 0 |
| Water | To 100% | To 100% |

20 Fluowet PL80 is 50:50 mix of perfluorinated alkyl phosphinic and perfluorinated alkyl phosphonic acid, supplied by CLARIANT. Antifoam MSA is based on polydimethylsiloxane supplied by Dow Corning.

EXAMPLE 2

The compositions of Example 1 (composition of the invention and comparison) were applied at the indicated rated expressed as glyphosate acid equivalent/ha to plant species grown in the glass-house. All treatments were made up in tap water and applied using a tracksprayer with a 11002 nozzle at a spray application volume of 2001/ha. All treatments were replicated 3 times. After spraying the plants were laid out in a glass-house and maintained at a temperature of 24°C by day and 19°C by night. A visual assessment of % control, where 0=unaffected and 100=complete kill, was carried out 22 days after treatment.

10

% Control Against Indicated Species

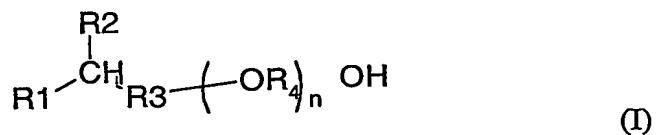
| Composition | Rate | AVEFA | BRODI | CHEAL | LOLRI | RAPRL |
|------------------------------|------|-------|-------|-------|-------|-------|
| Comparison | 240 | 32 | 3 | 57 | 7 | 20 |
| | 360 | 76 | 42 | 78 | 53 | 55 |
| | 540 | 86 | 87 | 92 | 73 | 85 |
| | 720 | 100 | 93 | 93 | 98 | 92 |
| Composition of the Invention | 240 | 48 | 15 | 87 | 25 | 58 |
| | 360 | 65 | 85 | 87 | 78 | 80 |
| | 540 | 62 | 100 | 92 | 95 | 73 |
| | 720 | 96 | 100 | 99 | 100 | 93 |

The species tested were:-

| | |
|----------|--|
| AVEFA | Wild Oat (<i>Avena fatua</i>) |
| BRODI | <i>Bromus diandrus</i> |
| 15 CHEAL | common lambsquarters (<i>Chenopodium album L.</i>) |
| LOLRI | <i>Lolium rigidum</i> |
| RAPRL | <i>Raphanus raphanistrum</i> |

CLAIMS

1. A herbicidal composition comprising glyphosate and a polyethyleneoxide alkanol having the formula



5 wherein R_1 and R_2 are methyl or ethyl, R_3 is a straight chain alkylene group containing from 5 to 12 carbon atoms, R_4 is an alkylene group containing 2 or 3 carbon atoms and n is from 4 to 20.

ABSTRACT

5

FORMULATION

10 A herbicidal composition comprising glyphosate and a poly(alkylene oxide) alkanol having the formula R₁R₂CH-R₃-(OR₄)_n-OH (I) wherein R₁ and R₂ are methyl or ethyl, R₃ is a straight chain alkylene group containing from 5 to 12 carbon atoms, R₄ is an alkylene group containing 2 or 3 carbon atoms and n is from 4 to 20. Especially preferred is ethoxylated, propoxylated or mixed alkoxyLATED/propoxylated iso-tridecyl alcohol wherein the degree of alkoxylation is from 6 to 14, for example the

15 commercially available product GENAPOL X080, iso-tridecyl alcohol having 8 polyethylene oxide units. The alkoxyLATED alcohol acts as both anti-foam and adjuvant despite being present at relatively low concentrations.